



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In the Application of: **SUZUKI, Noriyuki et al.**

Group Art Unit: 1714

Serial No.: 09/926,260

Examiner: **Katarzyna I. Wyrozebski Lee**

Filed: **October 3, 2001**

P.T.O. Confirmation No.: 8663

For. **THERMOPLASTIC RESIN COMPOSITION AND PROCESS FOR PREPARING THE SAME**

**SUBMISSION OF DECLARATION UNDER 37 CFR 1.132**

Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Date: September 21, 2004

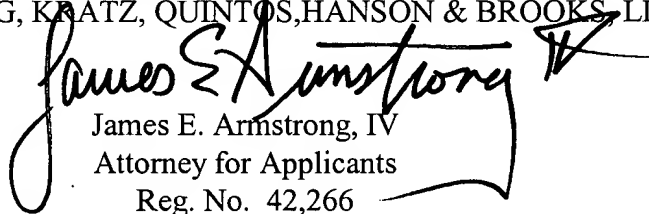
Sir:

Applicants respectfully submit a Declaration Under 37 CFR 1.132 discussed and stated in the Amendment timely filed on September 20, 2004, but accidentally not attached with the Amendment filed on September 20, 2004.

The Examiner's assistance in addressing this matter is greatly appreciated.

No fees are believed to be due. However, the Commissioner is authorized to charge any additional fees which may be required for consideration of this paper to Deposit Account No. 01-2340.

Respectfully submitted,  
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PATENT TRADEMARK OFFICE

Enclosure: Declaration Under CFR 1.132 (11 pages)

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE



In re the application of:

Noriyuki SUZUKI, et al.

Group Art Unit: 1714

Serial Number: 09/926,260

Examiner: Katarzyna Wyrozebski

Filed: October 3, 2001

For: THERMOPLASTIC RESIN COMPOSITION AND PROCESS FOR  
PREPARING THE SAME

DECLARATION UNDER 37 CFR 1.132

Commissioner for Patents  
Washington, D.C. 20006

Sir:

Noriyuki Suzuki residing 1-2-54 , Koyodai, Kawanishi-shi,  
Hyogo, Japan duly deposes and says:

1. That he graduated from Department of Synthetic  
Chemistry, Faculty of Engineering Science, OSAKA UNIVERSITY, Japan,  
in the year 1987.;

2. That since 1987, he has been employed in the capacity of  
KANEKA CORPORATION.;

3. That from 1987, he has been engaged in development for  
new polymer materials in the field of engineering plastics such as  
polyallylate, polyamide, polycarbonate and polyester.;

4. That he has read and is familiar with the instant  
application for United States Letters Patent and Office Action thereto  
mailed May 25, 2004.; and

5. That he has made experiments in order to show that a

polyester resin composition containing a thermoplastic polyester resin and layered phyllosilicate, which is prepared by the process comprising:

(A) a step for preparing a dispersion of layered phyllosilicate and water containing layered phyllosilicate and water;

(B) a step for drying the dispersion to obtain a powder of layered phyllosilicate;

(C) a step for mixing a component having low polymerization degree of the thermoplastic polyester resin with the powder of layered phyllosilicate; and

(D) a step for polymerizing the component having low polymerization degree of the thermoplastic polyester resin is inferior in physical property to the polyester resin composition obtained from Example 1 of the present specification.

6. Experiments were carried out by the following procedure.

#### Experiment 1

##### Step (A)

A dispersion of layered phyllosilicate and water was prepared by mixing 3,000 g of ion exchange-water and 160 g of Kunipia F (montmorillonite, basal space = 13 Å, available from Kunimine Industries Co., Ltd.) with stirring at 5,000 rpm for three minutes according to a wet mill (made by Nippon Seiki Co. Ltd.) and then 165 g of Ethoquad 18/25 (octadecylmethyl-[ethoxylated(15)] ammonium chloride, available from Lion Co. Ltd.) was added to the dispersion and fully blended.

##### Step (B)

The solids were then removed by filtration. Obtained filtercake was dried at 60 °C for 16 hours and then crushed into powder.

Step (C)

A reactor was charged with 3,000 g of Bishydroxyethyl terephthalate (NISSO-BHET available from Nisso Maruzen Chemicals Co., Ltd., hereinafter referred to as BHET), and BHET was melted under a flow of dry nitrogen with stirring at 140°C. Next, BHET was fully mixed with the powder of layered phyllosilicate obtained from Step (B).

Step (D)

Temperature of the system was elevated gradually while stirring. When the temperature reached about 220 to 240°C, to the system were added 7.0 g of a hindered phenol stabilizer (ADK STAB AO60 available from Asahi Denka Kogyo K.K.) and 0.6 g of antimony trioxide ( $\text{Sb}_2\text{O}_3$ ), and the system was gradually heated to 280°C. After heating, the system was depressurized (0.5 to 5.0 torr (0.0665 to 0.665MPa)) to carry out melt-polycondensation.

The above-obtained polyester resin composition was evaluated according to the following mannner (the same as in the present specification). Results are shown in Table 1.

(Logarithmic viscosity)

The thermoplastic resin composition of the present invention was dried (at 140°C for four hours), and then weighed precisely in amount of about 100 mg. Thereto was added 20 ml of a mixed solvent

of phenol/1,1,2,2-tetrachloroethane (1/1 in weight ratio), and it was dissolved at 120°C. Viscosity of solution was measured at 25°C for the PET type and at 20°C for the PBT type by using Ubbelohde's viscometer and an automatic measuring machine (Viscotimer made by Lauda Co., Ltd.). Logarithmic viscosity ( $\eta_{inh}$ ) was calculated from the following equation:

$$\eta_{inh} = \{ \ln (t / t_0) \} / C$$

(wherein  $t$  indicates time of solution,  $t_0$  indicates time of mixed solvent alone, and  $C$  indicates concentration (g/dl).

(Ash content ratio)

Ash content ratio of the thermoplastic resin derived from fibrous fiber and layered phyllosilicate was measured according to JIS K 7052.

(Measurement of dispersion state)

A micro thin piece having thickness of 50 to 100  $\mu\text{m}$  was used. The dispersion state of layered phyllosilicate was observed and photographed by using a transmission electron microscope (made by JEOL Ltd. JEM-1200) under accelerating voltage of 80kv at a magnification of X40,000 to 1,000,000. Then any region containing at least 100 particles was selected on an TEM image and layer thickness, layer length, the number of particles (value  $[N]$ ) and equivalent area circle diameter  $[D]$  were measured, manually by using a graduated ruler, or by processing according to an image analysis device PIASIII made by

Interquest Inc.

Average aspect ratio is defined as a number average value among the ratios of the layer length to the layer thickness of the layered phyllosilicate.

Value [N] was measured as follows. First, the number of particles of layered phyllosilicate present in the selected region was counted on the TEM image. Aside from this, ash content ratio of the resin composition derived from the layered phyllosilicate was measured. Value [N] was defined as a value obtained by dividing the above number of particles and converting it to the value for an area of  $100 \mu\text{m}^2$ .

The average layer thickness was defined as a mean value of thickness of each layered phyllosilicate, while the maximum layer thickness was defined as the largest value of thickness of each layered phyllosilicate.

When dispersion particles are too large to be observed by TEM properly, the value [N] was measured by using a light microscope (Optical Microscope BH-2 available from Olympus Co., Ltd.) in the same manner as the above. However, the sample was melted at 250 to 270°C, if necessary, by using Hot stage THM600 available from LINKAM Co., Ltd. and dispersion of particles kept in the state of melting was observed. The aspect ratio of dispersion particles which did not disperse in the thin plate-like shape was defined as the value of long diameter/short diameter. Herein, the long diameter refers to the long side of a rectangle which is the minimum in area among those circumscribed to target particles, while the minor axis refers to the short side of the above rectangle which is the minimum in area in a microscopic image and the like.

The equivalent area circle diameter [D] was measured by processing according to an image analysis device PIASIII made by Interquest Inc.

(Heat deformation temperature)

The thermoplastic resin composition was dried (at 140°C for five hours). A test specimen having a size of about 10 × 100 × 6 mm was prepared by injection molding under conditions of resin temperature of 250 to 280°C, gauge pressure of about 10 MPa and injection speed of about 50 % by using an injection molding machine whose clamping force is 75 t (IS-75E made by Toshiba Machine Co., Ltd.). Heat deformation temperature of the obtained test specimen was measured according to ASTM D-648 under load of 1.82 MPa. It can be said that the larger the value of the heat deformation temperature is, the better the heat resistance is.

(Flexural property)

Flexural strength and flexural modulus of a test specimen prepared in the same manner as in case of measuring heat deformation temperature were measured according to ASTM D-790. It can be said that the larger the value of flexural strength and flexural modulus are, the better the flexural properties are.

(Warpage)

The thermoplastic resin composition was dried (at 140°C for five hours), and then a plate-like test specimen having a size of about 120 × 120 × 1 mm was prepared by injection molding under conditions of

mold temperature of 50°C, resin temperature of 250 to 280°C, gauge pressure of about 10 MPa and injection speed of about 50 % by using an injection molding machine whose clamping force is 75 t (IS-75E made by Toshiba Kikai K.K.). The plate-like specimen was placed on a plane, and by pressing one corner of the four, the longest distance from the plane to one of the remaining three corners was measured by using a caliper and the like. The four corners were pressed respectively, and a mean value was calculated by the obtained warpage values. It can be said that the smaller the value of warpage is, the better the dimensional accuracy is.

(Coefficient of linear expansion (Anisotropy) )

JIS1 dumbbell specimen was used, which had thickness of about 3 mm and prepared under the same condition as in case of measuring heat deformation temperature. A piece about 7 mm × 7 mm in size was cut from the center part of the dumbbell specimen. It was kept at 20°C for five minutes by using SSC-5200 and TMA-120C made by Seiko Electron Co., Ltd., and then heated within a range of 20°C to 150°C at a temperature rising rate of 5°C/minute. Coefficient of linear expansion between 30 to 120°C was measured at MD direction and TD direction, and the ratio therebetween (MD/TD) was regarded as anisotropy. It can be said that the more the anisotropy approaches to 1, the better the dimensional accuracy is.

(Central linear average roughness)

Center line roughness was measured by using the above dumbbell specimen according to a surface roughness meter, surfcom



1500A made by Tokyo Seimitsu Co., Ltd. The smaller value thereof indicates that the surface is smoother.

(Mold releasing ability)

Injection molding was carried out under conditions of mold temperature of 120°C and resin temperature of 250 to 280°C by using an injection molding machine (made by Nikko Co., Ltd.) whose clamping force is 35 t and a mold for draft evaluation (a cylindrical, 30-mm long mold having diameter of 20 mm, specimen shape: draft angle = 1°). The mold releasing ability of the thermoplastic resin composition during the injection molding was evaluated according to the following criteria.

◎: Very good

○: Good

△: Poor

×: Bad

Table 1

	Example 1	Example 1 of the specification
Logarithmic viscosity (g/dl)	0.61	0.59
Ash content ratio (% by weight)	6.55	6.58
Ratio of [D] $\leq 3000 \text{ \AA}$ (%)	0	90
Average value of [D] ( $\text{\AA}$ )	21100	1090
Number of dispersed particles[N] (number/% by weight $\cdot 100\mu^2$ )	6.8	132
Average aspect ratio	3.2	143
Average layer thickness ( $\text{\AA}$ )	3100	75
Maximum layer thickness ( $\text{\AA}$ )	9300	300
Flexural strength (MPa)	98	125
Flexural modulus (MPa)	2890	6290
Heat deflection Temperature ( $^{\circ}\text{C}$ )	135	215
Warpage (mm)	Cannot be measured (*1)	<0.1
Coefficient of linear expansion ( $10^{-5}/^{\circ}\text{C}$ )	MD 8.01	5.62
	TD 12.36	5.63
Center line roughness (nm)	589	22
Mold releasing ability	$\times$	$\odot$

(\*1) : cannot be measured due to remarkable deformation of molded articles

## Results and Discussion

When a polyester resin composition containing a thermoplastic polyester resin and layered phyllosilicate is prepared by the process, wherein layered phyllosilicate is dispersed in water and then dried, thereby mixed in the powder form with the component having low polymerization degree of the thermoplastic polyester resin, layered phyllosilicate apparently exists as large agglomerate in the resin composition. Therefore, the obtained polyester resin composition is inferior in physical property such as flexural property and mold releasing ability to the polyester resin composition obtained from Example 1 of the present specification.

The undersigned declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

This 1st day of September, 2004

by Noriyuki Suzuki  
Noriyuki Suzuki

We, the undersigned witnesses, hereby acknowledge that Noriyuki Suzuki is personally known to us and did execute the foregoing Declaration in our presence on:

Date: September 1, 2004    Witness Yoshitaka Ono

Date: September 1, 2004    Witness Nagayoshi Adachi